

APPLICATION FOR UNITED STATES LETTERS PATENT

for

NESTED COMPACT FUEL PROCESSOR FOR PRODUCING A HYDROGEN
RICH GAS

by

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BACKGROUND OF THE INVENTION

Fuel cells provide electricity from chemical oxidation-reduction reactions and possess significant advantages over other forms of power generation in terms of cleanliness and efficiency. Typically, fuel cells employ hydrogen as the fuel and oxygen as the oxidizing agent. The power generation is proportional to the consumption rate of the reactants.

A significant disadvantage which inhibits the wider use of fuel cells is the lack of a widespread hydrogen infrastructure. Hydrogen has a relatively low volumetric energy density and is more difficult to store and transport than the hydrocarbon fuels currently used in most power generation systems. One way to overcome this difficulty is the use of reformers to convert the hydrocarbons to a hydrogen rich gas stream which can be used as a feed for fuel cells.

Hydrocarbon-based fuels, such as natural gas, LPG, gasoline, and diesel, require conversion processes to be used as fuel sources for most fuel cells. Current art uses multi-step processes combining an initial conversion process with several clean-up processes. The initial process is most often steam reforming (SR), autothermal reforming (ATR), catalytic partial oxidation (CPOX), or non-catalytic partial oxidation (POX). The clean-up processes are usually comprised of a combination of desulfurization, high temperature water-gas shift, low temperature water-gas shift, selective CO oxidation, or selective CO methanation. Alternative processes include hydrogen selective membrane reactors and filters.

Despite the above work, there remains a need for a simple unit for converting a hydrocarbon fuel to a hydrogen rich gas stream for use in conjunction with a fuel cell.

SUMMARY OF THE INVENTION

The present invention is generally directed to an apparatus and method for converting hydrocarbon fuel into a hydrogen rich gas. In one illustrative embodiment of the present invention, the apparatus for converting hydrocarbon fuel into hydrogen rich gas includes a plurality of modules arranged radially along a common axis. Such an arrangement permits a compact design and highly efficient heat transfer between differing reactor modules. Depending upon the reaction taking place within the module,

1 each module may include an annular layer of thermally insulating material disposed
2 between the shell and the respective processing core. In a similar manner, a module may
3 include a porous support member, such as screen, mesh, perforated plate, or porous
4 sintered plate. Such a porous support member may be included so as to support and
5 contain the contents of the module, particularly granular catalyst materials.

6 As noted above and described herein, the apparatus of the present invention is
7 used to carry out a series of reactions that converts hydrocarbon fuel to hydrogen rich
8 gas. In one illustrative embodiment of such an apparatus, the first module preferably has
9 a processing core that includes a partial oxidation catalyst or alternatively a steam
10 reforming catalyst or alternatively an autothermal reforming catalyst or alternatively
11 combinations and/or mixtures of such catalysts. A second module positioned adjacent to
12 the first module is preferably designed so that the processing core of the second module
13 includes a first heat exchanger. Such a heat exchanger may be a fin-typed heat
14 exchanger, tubing heat exchanger, heat pipe, or similar means that are capable of
15 exchanging heat.

16 Within the plurality of modules, a third module is preferably positioned adjacent
17 to the second module, the processing core of the third module including a desulfurizing
18 agent. A wide variety of desulfurizing agents may be used but preferably the
19 desulfurizing agent includes zinc oxide. A fourth module positioned adjacent to and in
20 fluid communication with the third module includes a processing core containing a water
21 gas shift catalyst. One of skill in the art should understand and appreciate that the water
22 gas shift catalyst may be a low temperature water gas shift catalyst that includes a
23 catalyst material selected from copper, copper oxide, zinc, platinum, rhenium, palladium,
24 rhodium, and gold and combinations and mixtures of these and similar materials.
25 Alternatively, the water gas shift catalyst is a high temperature water gas shift catalyst
26 that includes a catalyst material selected from ferric oxide, chromic oxide, copper, iron
27 silicide, platinum, palladium and other platinum group members, and combinations and
28 mixtures of these and similar materials.

29 The apparatus is further designed to include within the plurality of modules a fifth
30 module positioned adjacent to and in fluid communication with the fourth module. The
31 processing core of the fifth module includes a second heat exchanger. Such a heat

1 exchanger may include a fin-typed heat exchanger, tubing heat exchanger, heat pipe, or
2 similar means that are capable of exchanging heat.

3 The plurality of modules further includes a sixth module positioned adjacent to
4 the fifth module with which it is in fluid communication. The processing core of the sixth
5 module includes a carbon monoxide oxidation catalyst that preferably includes a material
6 selected from platinum, palladium, iron, chromium, manganese, iron oxide, chromium
7 oxide, manganese oxide, ruthenium, gold, cerium, lanthanum, and combinations and
8 mixtures of these and similar compounds.

9 The present invention also includes a process for converting hydrocarbon fuel into
10 a hydrogen rich gas. One such illustrative process utilizes the apparatus disclosed herein.
11 Such a process generally includes providing a fuel processor having a plurality of
12 modules arranged radially along a common axis, each forming an annular reaction
13 chamber. By feeding the hydrocarbon fuel successively through each of the above
14 described modules in an generally radial direction a hydrogen rich gas is produced in a
15 manner that optimizes space considerations and heat transfer considerations.

16

17 **BRIEF DESCRIPTION OF THE DRAWINGS**

18 The description is presented with reference to the accompanying drawings in
19 which:

20 FIG. 1 depicts a simple process flow diagram for one illustrative embodiment of
21 the present invention.

22 FIG. 2 depicts a first illustrative embodiment of a compact fuel processor
23 apparatus of the present invention; and

24 FIG. 3 depicts a second illustrative embodiment of a compact fuel psocessor
25 apparatus of the present invention.

26

27 **DESCRIPTION OF ILLUSTRATIVE EMBODIMENTS**

28 The present invention is generally directed to an apparatus for converting
29 hydrocarbon fuel into a hydrogen rich gas. In a preferred aspect, the apparatus and
30 method described herein relate to a compact processor for producing a hydrogen rich gas
31 stream from a hydrocarbon fuel for use in fuel cells. However, other possible uses are

1 contemplated for the apparatus and method described herein, including any use wherein a
2 hydrogen rich stream is desired. Accordingly, while the invention is described herein as
3 being used in conjunction with a fuel cell, the scope of the invention is not limited to such
4 use.

5 Each of the illustrative embodiments of the present invention describe a fuel
6 processor or a process for using such a fuel processor with the hydrocarbon fuel feed
7 being directed through the fuel processor. The hydrocarbon fuel may be liquid or gas at
8 ambient conditions as long as it can be vaporized. As used herein the term
9 "hydrocarbon" includes organic compounds having C-H bonds which are capable of
10 producing hydrogen from a partial oxidation or steam reforming reaction. The presence
11 of atoms other than carbon and hydrogen in the molecular structure of the compound is
12 not excluded. Thus, suitable fuels for use in the method and apparatus disclosed herein
13 include, but are not limited to hydrocarbon fuels such as natural gas, methane, ethane,
14 propane, butane, naphtha, gasoline, and diesel fuel, and alcohols such as methanol,
15 ethanol, propanol, and the like.

16 The fuel processor feeds include hydrocarbon fuel, oxygen, and water. The oxygen
17 can be in the form of air, enriched air, or substantially pure oxygen. The water
18 can be introduced as a liquid or vapor. The composition percentages of the feed
19 components are determined by the desired operating conditions, as discussed below.

20 The fuel processor effluent stream from of the present invention includes
21 hydrogen and carbon dioxide and can also include some water, unconverted
22 hydrocarbons, carbon monoxide, impurities (e.g. hydrogen sulfide and ammonia) and
23 inert components (e.g., nitrogen and argon, especially if air was a component of the feed
24 stream).

25 Figure 1 depicts a general process flow diagram illustrating the process steps
26 included in the illustrative embodiments of the present invention. One of skill in the art
27 should appreciate that a certain amount of progressive order is needed in the flow of the
28 reactants through the reactors disclosed herein.

29 Process step A is an autothermal reforming process in which two reactions, partial
30 oxidation (formula I, below) and optionally also steam reforming (formula II, below), are
31 combined to convert the feed stream F into a synthesis gas containing hydrogen and

1 carbon monoxide. Formulas I and II are exemplary reaction formulas wherein methane is
2 considered as the hydrocarbon:



5 The partial oxidation reaction occurs very quickly to the complete conversion of
6 oxygen added and produces heat. The steam reforming reaction occurs slower and
7 consumes heat. A higher concentration of oxygen in the feed stream favors partial
8 oxidation whereas a higher concentration of water vapor favors steam reforming.
9 Therefore, the ratios of oxygen to hydrocarbon and water to hydrocarbon become
10 characterizing parameters. These ratios affect the operating temperature and hydrogen
11 yield.

12 The operating temperature of the autothermal reforming step can range from
13 about 550°C to about 900°C, depending on the feed conditions and the catalyst. The
14 invention uses a catalyst bed of a partial oxidation catalyst with or without a steam
15 reforming catalyst. The catalyst may be in any form including pellets, spheres, extrudate,
16 monoliths, and the like. Partial oxidation catalysts should be well known to those with
17 skill in the art and are often comprised of noble metals such as platinum, palladium,
18 rhodium, and/or ruthenium on an alumina washcoat on a monolith, extrudate, pellet or
19 other support. Non-noble metals such as nickel or cobalt have been used. Other
20 washcoats such as titania, zirconia, silica, and magnesia have been cited in the literature.
21 Many additional materials such as lanthanum, cerium, and potassium have been cited in
22 the literature as “promoters” that improve the performance of the partial oxidation
23 catalyst.

24 Steam reforming catalysts should be known to those with skill in the art and can
25 include nickel with amounts of cobalt or a noble metal such as platinum, palladium,
26 rhodium, ruthenium, and/or iridium. The catalyst can be supported, for example, on
27 magnesia, alumina, silica, zirconia, or magnesium aluminate, singly or in combination.
28 Alternatively, the steam reforming catalyst can include nickel, preferably supported on
29 magnesia, alumina, silica, zirconia, or magnesium aluminate, singly or in combination,
30 promoted by an alkali metal such as potassium.

1 Process step B is a cooling step for cooling the synthesis gas stream from process
2 step A to a temperature of from about 200°C to about 600°C, preferably from about
3 300°C to about 500°C, and more preferably from about 375°C to about 425°C, to
4 optimize the temperature of the synthesis gas effluent for the next step. This cooling may
5 be achieved with heat sinks, heat pipes or heat exchangers depending upon the design
6 specifications and the need to recover / recycle the heat content of the gas stream. One
7 illustrative embodiment for step B is the use of a heat exchanger utilizing feed stream F
8 as the coolant circulated through the heat exchanger. The heat exchanger can be of any
9 suitable construction known to those with skill in the art including shell and tube, plate,
10 spiral, etc. Alternatively, or in addition thereto, cooling step B may be accomplished by
11 injecting additional feed components such as fuel, air or water. Water is preferred
12 because of its ability to absorb a large amount of heat as it is vaporized to steam. The
13 amounts of added components depend upon the degree of cooling desired and are readily
14 determined by those with skill in the art.

15 Process step C is a purifying step. One of the main impurities of the hydrocarbon
16 stream is sulfur, which is converted by the autothermal reforming step A to hydrogen
17 sulfide. The processing core used in process step C preferably includes zinc oxide and/or
18 other material capable of absorbing and converting hydrogen sulfide, and may include a
19 support (e.g., monolith, extrudate, pellet etc.). Desulfurization is accomplished by
20 converting the hydrogen sulfide to water in accordance with the following reaction
21 formula III:



22 Other impurities such as chlorides can also be removed. The reaction is
23 preferably carried out at a temperature of from about 300°C to about 500°C, and more
24 preferably from about 375°C to about 425°C. Zinc oxide is an effective hydrogen sulfide
25 absorbent over a wide range of temperatures from about 25°C to about 700°C and affords
26 great flexibility for optimizing the sequence of processing steps by appropriate selection
27 of operating temperature.

28 The effluent stream may then be sent to a mixing step D in which water is
29 optionally added to the gas stream. The addition of water lowers the temperature of the
30 reactant stream as it vaporizes and supplies more water for the water gas shift reaction of

1 process step E (discussed below). The water vapor and other effluent stream components
2 are mixed by being passed through a processing core of inert materials such as ceramic
3 beads or other similar materials that effectively mix and/or assist in the vaporization of
4 the water. Alternatively, any additional water can be introduced with feed, and the
5 mixing step can be repositioned to provide better mixing of the oxidant gas in the CO
6 oxidation step G disclosed below.

7 Process step E is a water gas shift reaction that converts carbon monoxide to
8 carbon dioxide in accordance with formula IV:



10 This is an important step because carbon monoxide, in addition to being highly
11 toxic to humans, is a poison to fuel cells. The concentration of carbon monoxide should
12 preferably be lowered to a level that can be tolerated by fuel cells, typically below 50
13 ppm. Generally, the water gas shift reaction can take place at temperatures of from
14 150°C to 600°C depending on the catalyst used. Under such conditions, most of the
15 carbon monoxide in the gas stream is converted in this step.

16 Low temperature shift catalysts operate at a range of from about 150°C to about
17 300°C and include for example, copper oxide, or copper supported on other transition
18 metal oxides such as zirconia, zinc supported on transition metal oxides or refractory
19 supports such as silica, alumina, zirconia, etc., or a noble metal such as platinum,
20 rhenium, palladium, rhodium or gold on a suitable support such as silica, alumina,
21 zirconia, and the like.

22 High temperature shift catalysts are preferably operated at temperatures ranging
23 from about 300° to about 600°C and can include transition metal oxides such as ferric
24 oxide or chromic oxide, and optionally including a promoter such as copper or iron
25 silicide. Also included, as high temperature shift catalysts are supported noble metals
26 such as supported platinum, palladium and/or other platinum group members.

27 The processing core utilized to carry out this step can include a packed bed of
28 high temperature or low temperature shift catalyst such as described above, or a
29 combination of both high temperature and low temperature shift catalysts. The process
30 should be operated at any temperature suitable for the water gas shift reaction, preferably
31 at a temperature of from 150°C to about 400°C depending on the type of catalyst used.

1 Optionally, a cooling element such as a cooling coil may be disposed in the processing
2 core of the shift reactor to lower the reaction temperature within the packed bed of
3 catalyst. Lower temperatures favor the conversion of carbon monoxide to carbon
4 dioxide. Also, a purification processing step C can be performed between high and low
5 shift conversions by providing separate steps for high temperature and low temperature
6 shift with a desulfurization module between the high and low temperature shift steps.

7 Process step F is a cooling step performed in one embodiment by a heat
8 exchanger. The heat exchanger can be of any suitable construction including shell and
9 tube, plate, spiral, etc. Alternatively a heat pipe or other form of heat sink may be
10 utilized. The goal of the heat exchanger is to reduce the temperature of the gas stream to
11 produce an effluent having a temperature preferably in the range of from about 90°C to
12 about 150°C.

13 Oxygen is added to the process in step F. The oxygen is consumed by the
14 reactions of process step G described below. The oxygen can be in the form of air,
15 enriched air, or substantially pure oxygen. The heat exchanger may by design provide
16 mixing of the air with the hydrogen rich gas. Alternatively, the embodiment of process
17 step D may be used to perform the mixing.

18 Process step G is an oxidation step wherein almost all of the remaining carbon
19 monoxide in the effluent stream is converted to carbon dioxide. The processing is carried
20 out in the presence of a catalyst for the oxidation of carbon monoxide and may be in any
21 suitable form, such as pellets, spheres, monolith, etc. Oxidation catalysts for carbon
22 monoxide are known and typically include noble metals (e.g., platinum, palladium)
23 and/or transition metals (e.g., iron, chromium, manganese), and/or compounds of noble
24 or transition metals, particularly oxides. A preferred oxidation catalyst is platinum on an
25 alumina washcoat. The washcoat may be applied to a monolith, extrudate, pellet or other
26 support. Additional materials such as cerium or lanthanum may be added to improve
27 performance. Many other formulations have been cited in the literature with some
28 practitioners claiming superior performance from rhodium or alumina catalysts.
29 Ruthenium, palladium, gold, and other materials have been cited in the literature as being
30 active for this use.

Two reactions occur in process step G: the desired oxidation of carbon monoxide (formula V) and the undesired oxidation of hydrogen (formula VI) as follows:



The preferential oxidation of carbon monoxide is favored by low temperatures. Since both reactions produce heat it may be advantageous to optionally include a cooling element such as a cooling coil disposed within the process. The operating temperature of process is preferably kept in the range of from about 90°C to about 150°C. Process step G preferably reduces the carbon monoxide level to less than 50 ppm, which is a suitable level for use in fuel cells, but one of skill in the art should appreciate that the present invention can be adapted to produce a hydrogen rich product with of higher and lower levels of carbon monoxide.

The effluent exiting the fuel processor is a hydrogen rich gas containing carbon dioxide and other constituents which may be present such as water, inert components (e.g., nitrogen, argon), residual hydrocarbon, etc. Product gas may be used as the feed for a fuel cell or for other applications where a hydrogen rich feed stream is desired. Optionally, product gas may be sent on to further processing, for example, to remove the carbon dioxide, water or other components.

In one illustrative embodiment of the present invention, the fuel processor is of modular construction with individual modular units, which are separable, rearrangeable, and individually replaceable. The modular units may be used in any orientation, e.g., vertical or horizontal orientation, and is adapted to be used in conjunction with a fuel cell such that the hydrogen rich product gas of the reactor described herein is supplied directly to a fuel cell as a feed stream. While the modules can have any cross sectional configuration, such as circular, rectangular, triangular, etc., a circular cross section is preferred with the overall reactor being of a generally tubular shape.

The modules are constructed in accordance with the description given below. The modules can be fabricated from any material capable of withstanding the operating conditions and chemical environment of the reactions described herein and can include, for example, stainless steel, Inconel, Incoloy, Hastelloy, and the like. The reaction pressure is preferable from about 0 to about 100 psig, although higher pressures may be

1 employed. The operating pressure of the fuel processor depends upon the delivery
2 pressure required by the fuel cell. For fuel cells operating in the 1 to 20 kW range an
3 operating pressure of 0 to about 100 psig is generally sufficient. The modules are of such
4 dimensions so as to safely contain the reaction at the desired operating pressures and
5 temperatures.

6 Depicted in Figure 2 is one illustrative embodiment of the present invention in
7 which a single stack of nested reactors is utilized as a fuel processor. Fuel feed stream
8 100 is preheated in a first heat exchanger 102 which is heated by the catalytic combustion
9 of the anode tail gas 104 generated by a fuel cell in the anode tail gas oxidation reactor
10 106. The catalytic combustion reaction is carried out in the anode tail gas oxidation
11 reactor using a suitable supported catalyst. Such supported catalysts oxidation are well
12 known in the art and may be in any suitable form, such as pellets, spheres, monolith, etc.
13 Oxidation catalysts for carbon monoxide are known and typically include noble metals
14 (e.g., platinum, palladium) and/or transition metals (e.g., iron, chromium, manganese),
15 and/or compounds of noble or transition metals, particularly oxides. A preferred
16 oxidation catalyst is platinum on an alumina washcoat. The washcoat may be applied to
17 a monolith, extrudate, pellet or other support. Additional materials such as cerium or
18 lanthanum may be added to improve performance. Many other formulations have been
19 cited in the literature with some practitioners claiming superior performance from
20 rhodium or alumina catalysts. Ruthenium, palladium, gold, and other materials have
21 been cited in the literature as being active for this use. Thus hot exhaust 108 generated
22 by the catalytic oxidation of the anode tail gas pre-heats the fuel to give a preheated fuel
23 feed 112. The pre-heated fuel feed is passed through a first desulfurization unit 114 so as
24 to substantially remove any sulfur containing compounds from the fuel. The desulfurized
25 fuel 116 is then combined with air or another suitable oxygen source 118 and water /
26 steam 120 to form a fuel / air / water mixture 122. The fuel / air / water mixture is
27 introduced into the reformation stack, distributed and pre-heated in the distribution
28 manifold. The preheated fuel / air / water mixture is introduced into the auto-thermal
29 reformation reactor 126 and undergoes reformation into a hydrogen containing gas. The
30 hydrogen containing gas is first cooled by a second heat exchanger 128 and then
31 desulfurized in the second desulfurization reactor 130. The resulting desulfurized

1 hydrogen containing gas is then subjected to a water-gas shift reaction in the shift reactor
2 132. The hydrogen containing gas exiting the shift reactor 132 is cooled by a third heat
3 exchanger 134 and then combined with an oxygen source, such as air source 138, and
4 then passed through a preferential oxidation reactor 136 so as to substantially remove
5 trace amounts of carbon monoxide that may be present in the hydrogen containing gas.
6 The hydrogen containing gas exiting the preferential oxidation reactor is subjected to
7 cooling in a fourth heat exchanger 140 before exiting the reactor as hydrogen rich gas
8 142. The hydrogen containing gas is preferably used in a fuel cell or may be stored or
9 used in other processes that should be apparent to one of skill in the art.

10 Upon careful examination of Figure 2, one of skill in the art should note that
11 except for the first desulfurization step, substantially all of the reactors in the fuel
12 processor are nested one inside the other. The order of nesting as illustrated is designed
13 so as to maximize the heat exchange between exothermic reactors and endothermic
14 reactors. Thus the nested reactors achieve an optimized thermal exchange balance that
15 optimizes the efficiency of the fuel processor in an overall compact area. In order to
16 start-up the fuel reformer, electric pre-heater 110 may be used to pre-heat anode tail gas
17 or some other suitable alternative startup fuel to initiate the reaction within the anode tail
18 gas oxidation reactor. It should also be noted that the general flow of reactants through
19 the fuel processor is proceeding from an inner core reactor to outer surrounding reactors
20 in a “radial manner”. A similar result may also be achieved by use of partially porous
21 reactor walls made of refractory materials.

22 One of skill in the art after reviewing the above description should understand and
23 appreciate that each module performs a separate operational function and is generally
24 configured as shown in Figure 2. Feed stream F (100) is introduced through inlet pipe
25 (not shown) and product gas P (142) is drawn off via outlet pipe (not shown). Module
26 126 is the autothermal-reforming module corresponding to process step A of Figure 1.
27 The autothermal-reforming module has built into it a heat exchanger 128 that cools the
28 outgoing product gas which corresponds to process step B of Figure 1. Module 130 is a
29 purifying module corresponding to process step C of Figure 1. Module 132 is a water gas
30 shift module corresponding to process step E of Figure 1. The cooling step corresponding
31 to process step F of Figure 1 is carried out by vertical finned heat exchanger 134.

1 Module 136 is a preferential oxidation step corresponding to process step G of Figure 1.
2 Air source 138 provides a source for oxygen to process gas for the oxidation reaction
3 (Equation V) of Module 136. Module 136 also contains a heat exchanger 140 (partially
4 shown) positioned within or surrounding the catalyst bed so as to maintain a desired
5 oxidation reaction temperature. One of skill in the art should appreciate that the process
6 configuration described in this embodiment may vary depending on numerous factors,
7 including but not limited to feedstock quality and required product quality.

8 Turning now to Figure 3, an alternative illustrative embodiment of a dual stack
9 fuel processor is shown in which there is a reforming stack 30 and a oxidizing stack 40.
10 As will become apparent below, the reforming stack 30 generally carries out the
11 reformation process in which fuel is converted into suitably pure hydrogen gas for use
12 with a fuel cell (not shown). The oxidation stack generally serves as a means for
13 recovering the heat content of the anode tail gas generated by the fuel cell and preheats
14 the fuel for the reforming stack.

15 Fuel 200 is provided to the reforming stack 40 that includes two major
16 components, the anode tail gas oxidation reactor 204 and the desulfurization reaction 210.
17 The anode tail gas oxidation reactor is designed such that it catalytically oxidizes the
18 anode tail gas 206 generated from a fuel cell. The heat generated is transferred to the fuel
19 200 by vertical fin heat exchangers 202 generating a cooled exhaust 208. The heated fuel
20 is then subjected to a desulfurization reactor 210 that substantially removes the sulfur
21 compounds present in the fuel. The desulfurized fuel 212 is provided to the reforming
22 stack 30 and combined with air 214 and hot water / steam 216. This mixture of fuel / air /
23 water passes through a distribution manifold 218 which directs the mixture through a
24 vertical fin heat exchanger 220. The pre heated fuel / air / water mixture is then
25 introduced into the auto-thermal reformation reactor 224. During start-up, an electrical
26 pre-heater element 222 is installed at the bottom of the auto-thermal reformation reactor.
27 However, once started the heat generated by the auto-thermal reformation reactor is
28 sufficient to heat the fuel / air / water mixture via the vertical finned heat exchangers 220.
29 The product gas from the auto-thermal reforming reactor is directed to a second
30 desulfurization reactor 226 which removes any remaining sulfur compounds from the gas
31 product stream. The desulfurized gas is then introduced into the water-gas shift reactor

1 228. The temperature of the shift reactor is controlled by tube heat exchangers 232. It
2 should be noted at this point that the heat exchanger in the present illustrative
3 embodiment is designed as one continuous tube routed throughout the reforming stack.
4 Points A and B are respectively connected to each other such that the flow of water
5 through the tube is continuous. After passing through the shift reactor, air 229 is mixed
6 with the hydrogen containing gas and passed to a preferential oxidation reactor 230. In
7 the preferential oxidation reactor trace amounts of carbon monoxide are removed so as to
8 purify the hydrogen containing gas for use in a fuel cell. The product gas if further
9 cooled by heat exchanger coils 232 and leaves the reforming stack as hydrogen
10 containing gas 236. The hydrogen containing gas is preferably used in a fuel cell or may
11 be stored or used in other processes that should be apparent to one of skill in the art.

12 One of skill in the art after reviewing the above description of Figure 3 should
13 understand and appreciate that each module performs a separate operational function.
14 Feed stream F (200) is introduced through inlet pipe (not shown) and product gas P (236)
15 is drawn off via outlet pipe (not shown). Module 224 is the autothermal reforming
16 module corresponding to process step A of Figure 1. The autothermal reforming module
17 has built into it a vertical fin heat exchanger the simultaneously heats the incoming fuel
18 mixture and cools the outgoing product gas which corresponds to process step B of
19 Figure 1. An electric heater 222, is installed at the bottom inlet of the autothermal
20 reformation reactor for start-up heat. Module 226 is a purifying module corresponding to
21 process step C of Figure 1. Module 228 is a water gas shift module corresponding to
22 process step E of Figure 1. The cooling step corresponding to process step F of Figure 1
23 is carried out by finned tube heat exchanger 232. In this illustrative embodiment, heat
24 exchanger 232 is shown as a general heat sink for the entire reforming stack, however,
25 one of skill in the art could redesign the heat exchangers to have multiple flows and heat
26 exchangers. Module 230 is an oxidation step corresponding to process step G of Figure
27 1. Air source 229 provides a source for oxygen to process gas for the oxidation reaction
28 (Equation V) of Module 230. Module 230 also contains a heat exchanger 232 (partially
29 shown) positioned within or surrounding the catalyst bed so as to maintain a desired
30 oxidation reaction temperature. One of skill in the art should appreciate that the process

1 configuration described in this embodiment may vary depending on numerous factors,
2 including but not limited to feedstock quality and required product quality.

3 In view of the above disclosure, one of ordinary skill in the art should understand
4 and appreciate that the present invention includes many possible illustrative embodiments
5 that depend upon design criteria. One such illustrative embodiment includes a compact
6 fuel processor for converting a hydrocarbon fuel feed into hydrogen rich gas, in which
7 the fuel processor assembly includes multiple concentric vessels for converting the
8 hydrocarbon fuel feed into the hydrogen rich gas. The hydrogen rich gas thus generated
9 is suitable for direct feed to a fuel cell or may be stored for use later or may be used in
10 processes or reactions that utilize hydrogen gas as a feed stock.

11 In such an illustrative embodiment, the processor assembly itself includes: an
12 oxidation core vessel containing an oxidation catalyst; a reforming vessel surrounding the
13 oxidation core vessel and forming a first annular space filled with autothermal reforming
14 catalyst; a desulfurizing vessel surrounding the reforming vessel and forming a second
15 annular space filled with desulfurization catalyst; a shift vessel surrounding the
16 desulfurizing vessel and forming a third annular space filled with water gas shift catalyst;
17 and a preferred oxidation vessel surrounding the shift vessel and forming a fourth annular
18 space filled with preferred oxidation catalyst. The above reactor areas, except for the
19 oxidation core vessel, are in fluid communication with each other so as to carry out the
20 desired fuel processing reactions needed to generate substantially pure hydrogen gas.
21 The oxidation core vessel oxidizes fuel cell anode tail gas or alternatively another
22 suitable fuel such as natural gas, to produce a hot exhaust gas. The hot exhaust gas in-
23 turn preheats the hydrocarbon fuel that is utilized in the fuel reformation reactions. In
24 one preferred illustrative embodiment, an electric heater is present for preheating the
25 anode tail gas prior to introducing the anode tail gas to the oxidation core vessel. The
26 illustrative compact fuel processor may include an external desulfurizing vessel for
27 desulfurizing the hydrocarbon fuel feed. Preferably the second desulfurizing vessel is a
28 replaceable canister so that it is serviceable without having to disassemble the nested fuel
29 processor stack. In the present illustrative embodiment, the hydrocarbon fuel feed is
30 sequentially introduced to the first annular space, then to the second annular space, then
31 to the third annular space, and then to the fourth annular space to produce the hydrogen

rich gas. A plurality of cooling coils for removing the heat of reaction produced in the first annular space, the second annular space, the third annular space, and the fourth annular space are preferably employed in the illustrative embodiment so as to efficiently distribute the heat generated by the exothermic reaction and the heat requirements of the endothermic reactions. A circulating coolant flows through the cooling coils and preferably the circulating coolant is selected from air, water, the hydrocarbon fuel feed, combinations and mixtures of these and similar items. In one preferred illustrative embodiment each annular space is surrounded by heat resisting refractory. Thus the heat transfer between annular spaces is controlled and the overall heat efficiency of the fuel processor can be optimized.

Yet another illustrative embodiment of the present invention is a compact fuel processor for converting a hydrocarbon fuel feed into hydrogen rich gas, that includes a heat exchanger coil; a reforming core vessel containing an autothermal reforming catalyst bed; a desulfurizing vessel surrounding the reforming core vessel and forming a first annular space filled with desulfurization catalyst; a shift vessel surrounding the desulfurizing vessel and forming a second annular space filled with water gas shift catalyst; and a preferred oxidation vessel surrounding the shift vessel and forming a third annular space filled with preferred oxidation catalyst. Such an illustrative compact fuel processor is designed such that the hydrocarbon fuel feed is preheated by the hydrogen rich gas in the heat exchanger coil to produce a preheated hydrocarbon fuel feed; and the preheated hydrocarbon fuel feed is sequentially introduced to the reforming core vessel, then to the second annular space, then to the third annular space, and then to the fourth annular space to produce the hydrogen rich gas.

It is preferred within this illustrative embodiment that the reforming core vessel has a set of external vertical fins for further preheating the preheated hydrocarbon fuel feed to produce a second preheated hydrocarbon fuel feed. Thus, the second preheated hydrocarbon fuel feed becomes the preheated hydrocarbon fuel feed introduced to the reforming core vessel. It should also be noted that the second annular space may have a second heat exchanger coil for reaction temperature control. Further the present illustrative embodiment can include an electrical heater for starting up the autothermal reforming catalyst bed.

1 While the apparatus, compositions and methods of this invention have been
2 described in terms of preferred or illustrative embodiments, it will be apparent to those of
3 skill in the art that variations may be applied to the process described herein without
4 departing from the concept and scope of the invention. All such similar substitutes and
5 modifications apparent to those skilled in the art are deemed to be within the scope and
6 concept of the invention as it is set out in the following claims.

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